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Positive muon Knight shift and spin relaxation in heavy fermion superconductors UPt_3 and UBe_{13}

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We report muon spin rotation/relaxation (μSR) measurements of the heavy fermion superconductors UBe_{13} and UPt_3. In both materials we find that the muon Knight shift is unchanged in the superconducting state, consistent with odd-parity pairing (such as p-wave). The magnetic field penetration depths in UPt_3 and UBe_{13} are extremely long, greater than 10000 Å. We find no evidence of a magnetic transition in UBe_{13} below 10 K.

Heavy fermion (HF) systems, materials where the conduction electrons have extremely large effective masses m^* (as exhibited in such properties as the electronic specific heat γ and the cyclotron mass) have been the subject of numerous investigations [#1]. Measurements of ultrasonic attenuation [2], magnetic field penetration depth [3], neutron scattering [4], NMR Knight shift [5,6] and specific heat [7] have provided evidence that the superconducting pairing in UBe_{13} and UPt_3 may be other than s-wave ($l=0$). The static spin susceptibility and the magnetic field penetration depth are two properties of the host system which reflect the symmetry of the superconducting state. Positive muon spin rotation/relaxation (μSR), through measurements of the muon Knight shift and the transverse field muon spin relaxation rate is a sensitive probe of these properties.

In an even-parity (such as s-wave) superconductor, the electrons are paired in states with opposite spin. Thus, the susceptibility of the pair is zero. Only states excited above the superconducting gap contribute to the spin susceptibility which falls from its normal state value to zero at $T=0$. The spin susceptibility χ_{SC} of an s-wave BCS superconductor is given in terms of the Yosida function $Y(T)$ as $\chi_{SC}=\chi_n Y(T)$ where χ_n is the normal state susceptibility. In real systems, spin–orbit scattering from surfaces or impurities can cause a reduction of this change in the susceptibility [8]. Two examples of triplet-paired states are the ABM and BW states (by analogy with superfluid ^3He). As a result of the different pairing symmetry, the susceptibility of these states are $\chi_{ABM}=\chi_n$ and $\chi_{BW}=\chi_n[\frac{2}{3}+\frac{1}{3}Y(T)]$. In either case, the spin susceptibility differs markedly from that of

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[#1] For a review of heavy fermion systems, see ref. [1].

a spin singlet superconductor; measurement of χ allows one to distinguish different pairing symmetries.

In a μSR experiment [9], muons are implanted one at a time and a histogram of muon-decay positrons is obtained as a function of the time after the muon arrival. The number of detected positrons is

$$N(t)=N_0\{B+e^{-t/\tau_\mu}[1-A_0\mathscr{P}(t)]\}, \quad (1)$$

where B is a time-independent background, $\tau_\mu=2.2$ μs is the muon lifetime, $A_0\sim0.3$ is the initial asymmetry and $\mathscr{P}(t)$ is the muon polarization in the direction of the detector. In transverse field experiments, the polarization function is $\mathscr{P}(t)=G_{xx}(t)\times\cos[\omega_\mu(t)+\phi]$ where $\omega_\mu=\gamma_\mu B_{loc}$ (γ_μ is the muon gyromagnetic ratio and B_{loc} is the local field experienced by the muon). The relaxation function $G_{xx}(t)$ is typically approximated by a Gaussian ($G_{xx}(t)=\exp(-\sigma^2t^2/2)$).

The flux lattice in the mixed state of a type-II superconductor provides an inhomogeneity ΔB in the magnetic field throughout the sample which (in high fields) is inversely proportional to the square of the penetration depth [10]. Since the relaxation rate σ is proportional to the field inhomogeneity, measurement of σ allows one to determine the penetration depth ($\sigma\propto1/\lambda^2$). The temperature dependence of the magnetic field penetration depth $\lambda(T)$ also provides information about the pairing symmetry. If there are no nodes in the gap, the penetration depth will show little temperature dependence as $T\to0$. If there are zeroes in the gap however, thermal pair-breaking will give a power law temperature dependence in the penetration depth at low temperatures [3].

The experiments were performed on the M15 surface muon channel at TRIUMF which provides a nearly 100% spin polarized beam of positive muons. The UPt_3 sample was a mosaic of pieces 3 mm in diameter and 0.35 mm thick, cut from a single crystal using spark erosion. It was mounted with the $\hat{c}$-axis along the direction of the muon beam, and along the direction of the main applied field. The polycrystalline UBe_{13} sample was approximately 1 cm in diameter and 2 mm thick. Both samples were glued to a pure silver backing attached to the copper sample holder of an Oxford Instruments model 400 top-loading dilution refrigerator. All of the measurements below T_c were made following field cooling, except where noted.

In HF materials, the Knight shift is large because of the large Pauli-like susceptibility of the heavy electrons. The local field B_{loc} is different from the applied field, with the difference being proportional to the applied field (since $K_\mu\propto\chi$). Fourier transforms of high transverse field (~4 kG) data taken in UPt_3 (and UBe_{13} discussed later) exhibit two signals, one from the sample and a smaller amplitude one from the silver backing. Since silver has a small, temperature-independent Knight shift, it provides an internal reference for the sample Knight shift. We find that the frequency shift (comprised of the Knight shift, Lorentz and demagnetizing shifts and a superconducting diamagnetic shift below T_c) of the UPt_3 signal is proportional to the applied field. Measurements at higher temperatures ($T<250$ K), show that the Knight shift, measured with the field along the $\hat{c}$-axis is proportional to the bulk susceptibility χ, also measured with the field along the $\hat{c}$-axis (and is not proportional to the susceptibility in the basal plane). This frequency shift at low temperatures (shown in fig. 1a), is temperature independent and in particular, there is no change below T_c. Just above T_c the

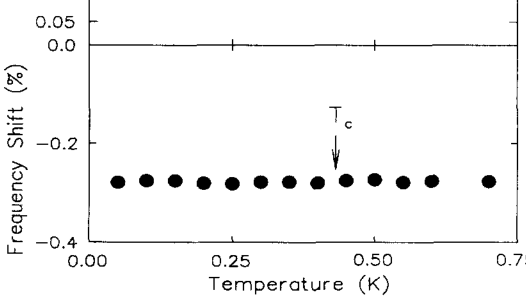

Fig. 1. Transverse field muon spin relaxation rate in UPt_3, measured in applied field $H_{ext}=3.9$ kG (filled circles) and $H_{ext}=150$ G (open circles). Inset shows field dependence of difference ($\Delta\sigma$) between relaxation as $T\to0$ and at $T>T_c$ (subtracted in quadrature). Lower portion of figure shows temperature dependence of the muon frequency shift. Lorentz and demagnetizing fields contribute about -0.12% of shift (determined in the normal state).

total shift is about -0.3%, Lorentz and demagnetizing fields (which, like the Knight shift are proportional to the spin susceptibility) contribute about -0.12% to this shift. The superconducting diamagnetic contribution to the frequency shift ($4\pi\chi_d(1-n)$) is extremely small, since the demagnetizing factor n is quite close to 1 in our geometry and χ_d is small in high fields.

We have measured the transverse field relaxation rate ($\sigma(T)$) going from the normal to the superconducting states as a function of external field for several fields $H\|\hat{c}$, up to 3.9 kG. At 3.9 kG (filled circles in fig. 1), there is at most an extremely small increase in the relaxation below $T_c \sim 0.4$ K. After subtracting the temperature-independent background, we find $\sigma(0) \leqslant 0.06\ \mu s^{-1}$, corresponding to a penetration depth $\lambda(0) \geqslant 11000$ Å.

Broholm et al. have reported transverse field μSR measurements [11] of UPt_3 in external fields somewhat less than 200 G. They extracted a low temperature penetration depth on the order of 8000 Å and discussed its temperature/orientation dependence in terms of d-wave superconductivity. Our results, measured in similarly low fields, are qualitatively consistent with those previously reported. With increasing field however, we find that the magnitude of the low temperature relaxation *decreases* (illustrated in the upper portion of fig. 1). The relaxation rate has not become field independent even by 3.9 kG as shown in the inset of fig. 1. A key result of the theory for extracting the penetration depth in the mixed state is that the field inhomogeneity ΔB should be independent of the field in a wide range of fields between H_{c1} and H_{c2} [10]. If the measured inhomogeneity is field dependent it generally implies that the measured relaxation does not accurately reflect the penetration depth. In this case, the value of 11000 Å can only serve as a *lower* bound on the penetration depth which may in fact be much longer.

There are several possible sources of increased relaxation in low fields. One of these is flux pinning, acting to prevent the formation of a uniform flux lattice. This is plausible, in view of the greatly enhanced relaxation observed in zero field cooled measurements (with $H_{ext}=3.9$ kG), which is characteristic of strong flux pinning. Shape-dependent inhomogeneities in the demagnetization factor can also provide broadening of the local field, roughly proportional to the sample magnetization ($-4\pi M$), which decreases with increasing field above the lower critical field H_{c1}[#2].

Several experiments (see for example ref. [14]) have detected an anomaly in UPt_3 around $H=12$ kG (for $B\|\hat{c}$). It has been suggested that such an anomaly may indicate a possible phase boundary between different superconducting states above and below this field. There is a possibility that this feature may be related to our observed field dependence in the relaxation rate. It is, however, not possible to assess the feasibility or magnitude of this effect, due to a lack of theoretical understanding of the superconducting states in UPt_3. Since the boundary to the normal state is at much higher field ($H_{c2}(0) \sim 2$ T), in general, we do not expect much change in the intrinsic penetration depth with the field $H_{ext} < 4$ kG.

UBe_{13} also possesses a large Knight shift, giving a frequency shift of -0.27% just above T_c. Lorentz and demagnetizing fields contribute -0.09% to this shift. In 3.5 kG, we see that the frequency shift (shown in fig. 2) decreases slightly below T_c. This corresponds to a change in the local field at the muon site of about 1 G. This increase in frequency however occurs with the same absolute value $B_{loc}(T\to 0) - B_{loc}(T\sim T_c) \approx 1$ G over the range 50 G $\leqslant H_{ext} \leqslant 3.5$ kG, independent of H_{ext} (see inset of fig. 2). After removing the field-independent frequency shift, we see that the Knight shift itself is essentially the same in the superconducting and normal states.

We note that the positive change in the precession frequency even in fields as small as 50 or 100 G, means that the local field at the muon site is actually significantly *larger* than in the normal state at these low fields. The source of this small additional field is not yet understood, although it is clearly correlated with the superconductivity of this system (since it appears at T_c). Previous high field μSR measurements of Heffner et al. [15] observed a larger fre-

[#2] Shivaram et al. [12] and de Visser et al. [13] measured $H_{c1} \sim 150$–200 G in UPt_3 using rf field penetration and dc-magnetization techniques respectively. Typically $B_{ext} > 5H_{c1}$ is necessary to ensure a uniform flux lattice. If their techniques give correct values for H_{c1}, then 200 G is clearly inadequate for determining $\lambda(T)$. However, thermodynamic arguments give values for H_{c1} of several gauss. In an applied field of only 20 G we do not observe any flux expulsion. This is consistent with either strong flux pinning or a small H_{c1}.

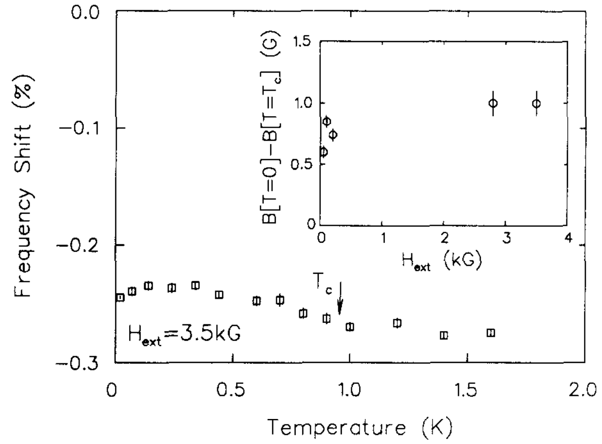

Fig. 2. Muon frequency shift in UBe_{13}, measured in $H_{ext}=3.5$ kG. Inset shows that the magnitude of frequency shift is largely independent of the applied field, meaning that it does *not* reflect the spin susceptibility. The Knight shift itself is essentially unchanged in the superconducting state. Lorentz and demagnetizing fields contribute about −0.09% to the shift (determined in the normal state).

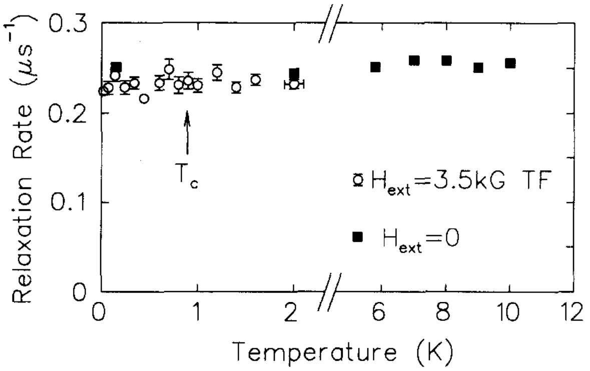

Fig. 3. Muon spin relaxation rate in UBe_{13}, measured in a transverse field of 3.5 kG (circles), and zero field (squares). The absence of an increase in the transverse field relaxation below T_c indicates that the penetration depth is greater than 10000 Å. The temperature independence of the zero field relaxation means that the local field at the muon site due to possible magnetic order must be less than 0.5 G.

quency shift than we report. We do not fully understand the origin of the difference; the different geometry (giving $n\sim0.15$ versus $n\sim0.8$ in our measurements) might contribute to the difference. We note that zero field measurements (see fig. 3) show no change above and below T_c, indicating the extra local field does not result from static magnetic order.

Results of high transverse field (4 kG) relaxation rate measurements above and below the superconducting transition temperature $T_c=0.9$ K are shown in fig. 3. The absence of any increase below T_c indicates that the penetration depth is greater than 10000 Å in UBe_{13}.

Recent magnetostriction results [16] have indicated that a transition, which was assumed to be to an antiferromagnetic state occurs in pure UBe_{13} with $T_n=8.8$ K. The sensitivity of μSR to ordering with very small moments has been demonstrated in UPt_3 [17] ($\mu\sim0.001\mu_B$) $CeCu_2Si_2$ [18] and URu_2Si_2 [19,20] ($\mu=0.03\mu_B$) which all have coexisting superconducting and magnetically ordered ground states. We have performed both zero and transverse field μSR measurements in UBe_{13} in the temperature range 20 mK$<T<$10 K. The absence of any change in the relaxation rates with temperature implies that the local field from magnetic order below 10 K at the muon site must be smaller than 0.5 G. Except for the unlikely possibility that the muon occupies a site in UBe_{13} where the local field is zero by symmetry, this result shows that there is no magnetic order within our sensitivity.

Both UPt_3 and UBe_{13} have muon Knight shifts which are unchanged from the normal to superconducting states. These results are in agreement with ^{195}Pt, ^{9}Be NMR [6,5] measurements of powdered samples as well as induced moment form factor measurements [4] using single crystals. Our new results for UBe_{13} have removed the inconsistency between μSR and these other techniques for the spin susceptibility. The observation of a temperature independent Knight shift is consistent with odd-parity (most likely p-wave) pairing. If the Knight shift was purely of orbital origin, we would not expect any effect below T_c. However we expect that there should be a large Pauli spin susceptibility in these materials in view of the large effective masses. In addition, the ratios of the susceptibility to the specific heat coefficient χ/γ are close to those of simple metals, as would be expected for a Fermi liquid, arguing against a purely orbital susceptibility. More theoretical and experimental work will be required to reconcile this result with the prevailing picture of d-wave (even parity) pairing in UPt_3.

Although it is possible for spin–orbit scattering to reduce the change in the Knight shift we would argue that this is not the case for these measurements. The mean free path in UPt_3 is greater than 1000 Å [21] which puts this material clearly in the clean limit. The use of powder samples in NMR measurements has led to suggestions of surface scattering; since μSR experiments do not require rf sample penetration, bulk samples are used, which should avoid significant surface scattering.

In both compounds we find that the penetration depths are in excess of 10000 Å. Flux confinement measurements [22] of the penetration depth have given a value of 11000 ± 2000 Å and 19000 ± 2000 Å for the low temperature value $\lambda(0)$ for UBe_{13} and UPt_3, respectively, which are consistent with the limits we can set with μSR. The penetration depth is given in terms of the carrier density and the carrier effective mass ($\lambda^2=m^*c^2/4\pi n_s e^2$). Effective masses are large in these systems (for example the cyclotron effective mass $m_c=25\rightarrow 90m_e$ in UPt_3 [21]) and so we expect that the penetration depths should be long in heavy fermion systems. Indeed, we have obtained shorter values for λ in systems with lighter effective masses such as URu_2Si_2 (8600 Å) [20] and U_6Fe (3200 Å) [20]. Since the low field relaxation in UPt_3 most likely does not directly reflect the penetration depth, we cannot discuss its temperature/orientation dependence as evidence for any particular pairing symmetry.

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